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MASTER

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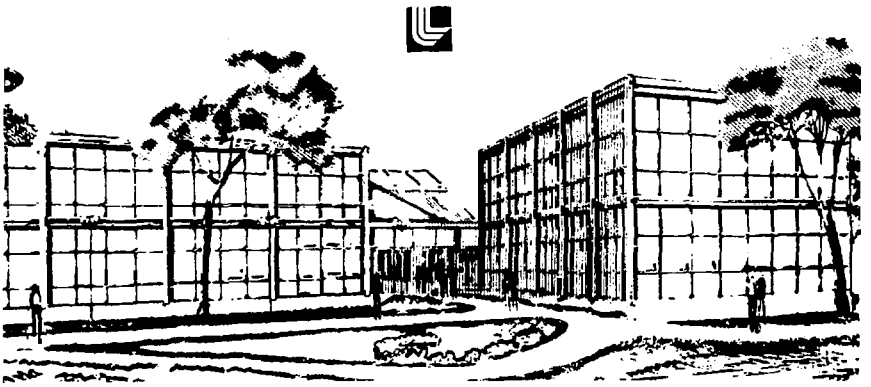

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EVALUATION OF THE USE OF SLUDGE CONTAINING PLUTONIUM AS A SOIL CONDITIONER FOR FOOD CROPS*

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INTRODUCTION

During the three-week period from 25 May to 16 June 1967, approximately 32 mCi of ^{239}Pu was inadvertently released from Lawrence Livermore Laboratory into the sanitary sewer system. It was concluded at the time from an investigation conducted by LLL in cooperation with ERDA-SAN and the California Department of Public Health that the release did not present a hazard to treatment plant personnel or the community.

The quantity of ^{239}Pu released was determined from the analysis of the samples collected by the Laboratory's sewage sampling system. This system collects a sample which is proportional in volume to the rate of flow in the sewer.

Analysis of the liquid collected by the Laboratory's sewage sampling system indicated that the maximum daily average concentration of the release did not exceed 2.1×10^{-6} μCi/ml. It is not known whether the chemical form of the ^{239}Pu was soluble or insoluble. The MPC for soluble ^{239}Pu, which is more restrictive than for insoluble ^{239}Pu, is 1×10^{-7} μCi/ml.

Immediately following the release, a detailed sampling program was begun at the Livermore sewage treatment plant. The results of this sampling program confirmed that approximately 30 mCi of ^{239}Pu had been released and that the ^{239}Pu was essentially all contained in the digested sludge at the sewage treatment plant. This digested sludge is accumulated in holding lagoons and transferred to drying beds each year during the summer. Sludge taken from the drying beds in recent years has a ^{239}Pu contamination level of about 2.3×10^{-6} μCi/g (dry weight).

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Large quantities of the digested sludge from the Livermore sewage treatment plant are used by municipal agencies as a soil conditioner in parks and in landscaping around public buildings. The dried digested sludge is also available without cost to the general public, and is commonly used as a soil conditioner for home lawns and gardens.

In view of this widespread use of the sludge as a soil conditioner, a more recent evaluation was made to confirm the original assessment of the possible adverse health implications associated with the use of the sludge. Two potentially significant uptake pathways were studied: (1) the resuspension and inhalation of the ^{239}Pu when the sludge is being used as a soil conditioner, and (2) uptake of the ^{239}Pu by plants grown in the sludge and its subsequent ingestion by people eating the crops. This paper presents the results of these two investigations.

EXPERIMENTAL METHODS

Soil Preparation

Two garden plots 10 m x 17 m each were established in the northeast corner of the LLL site to provide control and experimental conditions for the study. The soil at the garden site is classified as San Ysidro loam [1], and is common to the more level portions of the Livermore Valley. The soil is characterized as being medium acid with a silt-to-fine-sandy-loam texture and is used principally for pasture, range, grain, and grain hay.

Approximately 13 m^3 of the dried sludge was spread over the experimental garden plot, yielding a sludge depth of from 7.5 to 10 cm. The area was allowed to dry four weeks before being rototilled.

During the drying period there was no measurable precipitation. The average daytime maximum temperature during this interval was 16°C, and the average relative humidity as measured between 2 and 4 p.m. each afternoon was 40%. At the conclusion of the four-week drying period, the consistency of the sludge was dry and granular.

Samples of the sludge were taken for analysis of chemical makeup (Table I). The average ^{239}Pu content was 2.8×10^{-6} μCi/g dry weight with a range from 2.2 to 4.4×10^{-6} μCi/g. Chemical content of the sludge was found to be within the normal range found in municipal sludges with some minor variations.

After the drying period, the soil was rototilled to a soil depth of about 14 cm. The control garden was similarly rototilled, but without the addition of soil conditioner. The rototilling apparatus was 2 m wide and was pulled behind a tractor. The dust cover on the rototiller, which is normally closed during operation to minimize dust and flying dirt, was opened to allow the maximum amount of dust to escape. The rototilling operation, which lasted approximately 20 min, generated a large dust cloud. Core samples to a depth of 25 cm were taken from the two gardens. Portions of the core were taken and analyzed for plutonium. Analysis of the samples indicated the radioisotope content (dry-weight basis) given in Table II.

Plutonium content of the control soil is within the range of values for plutonium deposited by global fallout found in the Livermore-Amador Valley [2].

Dust Cloud Sampling

The dust cloud generated by the rototilling operation was carried by a 2-m/s wind over a high-volume Anderson cascade impactor as shown in Fig. 1. The Anderson cascade impactor samples at a rate of 0.6 m^3/min and consists of four collection stages with particle size collection characteristics shown in Fig. 2. Following the four collection stages is a backup filter whose purpose is to collect those submicron particles which are not

retained on any of the collection stages. The arbitrarily defined lower cutoff on each stage is the size at which 50% of the particles with that aerodynamic diameter pass on to the next smaller stage. The effective aerodynamic particle size collection range for each of the high-volume cascade impactor stages is shown in Table III.

Plutonium Solubility

To calculate the radiation dose to an individual exposed to airborne or ingested plutonium it is necessary to determine whether the chemical form of the plutonium is soluble or insoluble in body fluids. The dissolution characteristics of the plutonium in the sludge were determined using a technique developed by Kanapilly et al. [3] at the Lovelace Foundation. The basic apparatus used is a parallel-flow dissolution system, a schematic of which is shown in Fig. 3. It consists primarily of a dissolution chamber which contains the sample to be studied and a filter paper to physically separate the sample from the solvent. As the solvent passes the sample, it elutes any soluble material diffusing through the filter. The filter used was a Nucleopore type which has a discrete 0.1 μ pore size. Ten grams of the ^{239}Pu-contaminated sludge with a total activity of 2.36 x 10^{-5} μCi of ^{239}Pu were placed in the dissolution chamber. The solvent is formulated to simulate blood serum, and its composition is given in Table IV. The serum simulant was pumped through the dissolution chamber at a rate of 0.5 ml/min and collected for analysis. The collected solvent fractions were analyzed after 1, 2, 3, 6, 9, and 13 days. During the actual dissolution study with the ^{239}Pu-contaminated sludge, the discoloration of the solvent from material diffusing through the filter was readily visible.

Food Crop Production

Nineteen varieties of vegetables were selected for planting that were representative of seed, root, stem, flower, and fruit types grown in home gardens in the Livermore area. Those that provided sufficient mass for plutonium measurement included corn, turnips, broccoli, tomatoes, cucumbers and melons. Each plot was automatically sprinkler-irrigated approximately one-half hour each day to assure that soil moisture content approached field capacity. Seeds were planted in rows about 15 m long. The vegetables were harvested as they matured and were cleaned using techniques that approximated those a home gardener would use. For example, turnip roots were subjected to vigorous scrubbing in water to remove dirt. Also, parts of the turnip damaged by insect penetration were excised. The cleaned roots were freeze-dried to remove moisture and then were ground. The other vegetables were treated similarly, except that the corn was oven-dried to facilitate kernel removal.

Fifty to one hundred grams of each prepared vegetable were chemically digested and counted using alpha spectrometry to determine ^{239}Pu content. Samples of these vegetables plus the others which yielded smaller masses were analyzed by neutron activation and atomic adsorption techniques for trace-element uptake. Results of the trace metal experiment have been reported elsewhere [4].

RESULTS

Resuspension

The analysis of the resuspended dust collected by the high-volume cascade impactor is presented in Table III. The total ^{239}Pu activity collected by the cascade impactor was 3.0 x 10^{-7} μCi. At least 54% of the resuspended ^{239}Pu was collected on the submicron filter which follows the

fourth impactor collection stage. Thus, the majority of the resuspended ^{239}Pu was associated with particles having aerodynamic diameters less than 1.1 μ. Also shown in Table III are the approximate percentage depositions in the pulmonary region of the lungs [5] for the aerodynamic particle sizes associated with each cascade impactor stage. When these approximate deposition factors are multiplied by the activity collected on each of the impactor stages and the results are summed, the total is 7.8×10^{-8} μCi. Thus, the fraction of the total activity collected by the impactor which if inhaled would result in pulmonary deposition is $7.8 \times 10^{-8}/3.0 \times 10^{-7}$ or 0.26.

The results of the study to determine the solubility of the ^{239}Pu indicated that the ^{239}Pu is very insoluble in biological fluids. The fractional dissolution rates per day ranged from 8.3×10^{-7} to $\leq 1.4 \times 10^{-6}$, and are listed in Table V. The average dissolution rate per day for the 13-day period was $\leq 8.5 \times 10^{-7}$ d^{-1}. This value is in good agreement with the fractional dissolution rate obtained by Kanapilly et al. [3] for monodisperse $^{239}PuO_2$ particles. The physical diameter of the $^{239}PuO_2$ particles they used was 0.66 μ, and they studied the dissolution rate over a three-week period.

The average concentration of resuspended ^{239}Pu in the dust cloud as measured by the high-volume cascade impactor during the rototilling operation was 2.4×10^{-8} μCi/m^3. This concentration is less than 2.5% of the MPC_a for insoluble ^{239}Pu for exposure to a member of the general public.

Based on the data from the cascade impactor and the results of the solubility study, the maximum organ burdens and integrated organ doses that could result from the resuspension of the contaminated sludge when used as a soil conditioner were calculated. The calculations were based on the following data and assumptions:

1. The airborne concentration of the ^{239}Pu was assumed to be 2.4×10^{-7} μCi/m^3. This is 10 times the concentration measured by the high-volume cascade impactor. Although the ^{239}Pu concentration in the breathing zone of an individual adjacent to the rototiller would be somewhat higher than that measured by the cascade impactor, we believe that this factor of 10 is sufficiently conservative.
2. The aerodynamic particle size distribution used is that given in Table III. The effective aerodynamic particle size of the ^{239}Pu-contaminated dust on the submicron filter was assumed to be 0.5 μ.
3. The duration of the exposure was assumed to be 4 hours.
4. The chemical form of the ^{239}Pu is insoluble. The effective elimination half-life for insoluble ^{239}Pu in the pulmonary region of the lungs based on the revised ICRP Lung Model is 500 days [5].

The calculation was made using the revised ICRP Lung Model. This lung model has recently been computerized [6] to simplify calculations.

The results of the calculations are shown in Figs. 4 and 5. The pulmonary, bone, liver, and pulmonary lymph node burdens as a function of time are shown in Fig. 4. The integrated organ doses as a function of time are shown in Fig. 5. The organ burdens and integrated doses were calculated for a period of 50 years following the hypothetical intake.

The pulmonary lymph nodes received the largest integrated 50-year dose of approximately 2.7 mrad or 27 mrem. However, due to the apparent insensitivity and relative physiological unimportance of the pulmonary lymph nodes, the lungs are considered to be the critical organ for exposure to insoluble ^{239}Pu [7]. The total integrated 50-year dose to the pulmonary

region of the lungs is approximately 0.6 mrem. The total integrated dose to the bone is approximately 1.2 mrem. The total integrated dose to the liver is approximately 0.55 mrem.

Food Crop Uptake

Plutonium concentrations (dry-weight basis) in the sludge and control garden vegetables are shown in Table VI.

It would appear from the data that the control garden vegetables picked up a higher proportion (about 100 times) of the available plutonium in the control soil than did the vegetables grown in the sludge-amended soil. The actual causes of this apparent inconsistency were not determined, but may be due to different plutonium forms in the two soils, organic or other chemical complexing of the plutonium in the sludge, and the presence of high concentrations of other multivalent cations in the sludge. Another possible explanation is that plants discriminate against elevated plutonium concentrations. Also, for the sludge-amended garden, the apparent discrimination factors for plutonium uptake in the vegetables ranged from 2.9×10^{-5} for the corn seed to 1.6×10^{-4} for the broccoli. This may be due to the larger exposed surface area of the broccoli stem and flower and incomplete removal of soil particles containing elevated plutonium. In any event, these are considerations that deserve further study.

The radiation dose an individual would receive by consuming the sludge-garden vegetables was calculated using methods outlined in ICRP Publication 2 for evaluation of dose to internal organs due to ingestion of contaminated material [8]. Maximum organ burdens for the liver and bone were calculated based on the following assumptions:

1. Plutonium in the vegetation is soluble in biological fluids.
2. Plutonium concentration in the vegetation is 6.8×10^{-11} μCi/g.
3. A hypothetical person consumes 400 g/day [9] (wet weight), or 80 g/day (dry weight) of vegetables over a 50-year period.
4. Absorption fractions, transfer coefficients, and other plutonium factors are as given in ICRP-2 and ICRP-19 [8,10].

Under these assumptions, the total integrated doses to the critical organs are:

Organ	TID_{50}	Fraction of guide value [11]
Bone	2.2×10^{-5} rem	1.5×10^{-7}
Liver	1.5×10^{-5} rem	2.6×10^{-7}

These doses represent a very small fraction of recommended limits for non-occupational exposures.

CONCLUSIONS

This experiment was conducted to assess the potential hazard associated with the use of sludge containing plutonium as a soil conditioner for food crops.

Conditions were chosen that would maximize exposure to the ^{239}Pu in the sludge through resuspension and in plant content and thus approximated the maximum potential hazards due to the inhalation and ingestion pathways.

The estimated 50-year radiation doses to the pulmonary region of the lung, bone, and liver based on the results of the inhalation experiment are 6×10^{-4} rem, 1.2×10^{-3} rem, and 0.55×10^{-4} rem, respectively. Similarly, the 50-year radiation doses attributable to ingestion of the sludge-grown vegetables were 2.2×10^{-5} rem to the bone and 1.5×10^{-5} rem to the liver. Thus, the inhalation pathway is the more critical of the two. The maximum

permissible annual doses to the lungs, bone, and the liver for a member of the general public are 1.5, 3.0, and 1.5 rem, respectively [11]. Thus, the maximum credible 50-year lung, bone, and liver dose commitments associated with the use of the ^{239}Pu-contaminated sludge as a soil conditioner are approximately 4.0×10^{-2}% of the annual maximum permissible dose.

Under more realistic exposure circumstances, one might expect less drying of the sludge, less resuspension of dust and flying dirt before and during rototilling, and a much smaller sludge vegetable consumption rate. The conservative assumptions made in this analysis tend to assure that actual radiation doses would be even less than those calculated.

ACKNOWLEDGMENT

The authors wish to acknowledge the contributions of John H. Rehder and Thomas Todachine in planting, maintaining and harvesting of the test gardens. Similarly, we wish to thank Jesse W. Meadows for providing the radiochemical analysis of the samples.

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TABLE I. Chemical makeup of sludge.

Chemical Analysis (Dry weight basis)	
Ash	59.62%
Organic matter	40.38%
Organic nitrogen (N)	2.52%
Ammonia nitrogen (NH_3-N)	0.005%
Phosphorus (P)	1.72%
Potassium (K)	0.18%
Calcium (Ca)	2.56%
Iron (Fe)	1.68%
Magnesium (Mg)	0.74%
Aluminum (Al)	0.84%
Sulfate sulfur (SO_4-S)	0.22%
Manganese (Mn)	230 ppm
Fluorine (F)	35 ppm
Arsenic (As)	8 ppm
Copper (Cu)	475 ppm
Sodium (Na)	1,060 ppm

TABLE 11. Control and sludge-amended soil ^{239}Pu concentrations.

Sample	^{239}Pu (μCi/g)	Error (2σ)
Control soil	2.40×10^{-9}	6.8%
Sludge-amended soil	4.30×10^{-7}	2.7%

TABLE III. Results from high volume cascade impactor and projected pulmonary deposition.

Cascade impactor state	Aerodynamic particle size range (μ)	^{239}Pu, ^{240}Pu μCi on stage	Approximate % pulmonary deposition	μCi deposited in pulmonary region of the lungs[c]
1	>7	1.5×10^{-8}	10	1.5×10^{-9}
2	3.3 - 7	1.3×10^{-8}	15	2.0×10^{-9}
3	2.0 - 3.3	1.7×10^{-8}	20	3.4×10^{-9}
4	1.1 - 2	$<9.0 \times 10^{-8}$ [b]	25	$<2.3 \times 10^{-8}$
Submicron Filter	<1.1[a]	1.6×10^{-7}	30	4.8×10^{-8}
	TOTAL	3.0×10^{-7}		7.8×10^{-8}

a The aerodynamic particle size for the plutonium collected on the filter was assumed to be 0.5 μ.

b The minimum detectable activity on stage 4 was relatively high due to a problem with chemical recovery efficiency during the processing of the sample.

c The activity on each stage that would be deposited in pulmonary region of lungs if an individual had inhaled the same air as sampled by the cascade impactor.

TABLE IV. Constituents in serum simulant[a]

Chemical Compound	m moles/liter of distilled H_2O
NaCl	116
NH_4Cl	10
$NaHCO_3$	27
NaH_2PO_4	1.2
Na_3 Citrate	0.2
Glycine	5.0
l-cysteine	1.0
H_2SO_4	0.5
$CaCl_2$	0.2

a Reference 3.

TABLE V. Dissolution rates for ^{239}Pu in sewer sludge.

Sample	Day	μCi dissolved per minute	Fraction dissolved per day[a]
1	1	$<2.3 \times 10^{-14}$	$<1.4 \times 10^{-6}$
2	2	1.8×10^{-14}	1.1×10^{-6}
3	3	1.8×10^{-14}	1.1×10^{-6}
4	4-6	$<1.4 \times 10^{-14}$	$<8.3 \times 10^{-7}$
5	7-9	1.4×10^{-14}	8.3×10^{-7}
6	10-13	1.4×10^{-14}	8.3×10^{-7}

a The original sample contained 10 grams of sludge with a total ^{239}Pu activity of 2.3×10^{-5} μCi.

TABLE VI. Plutonium concentrations in sludge-garden vegetables.

Vegetable	Soil	^{239}Pu ($\mu Ci/g$)	Error (2σ)	Plutonium plant/soil ratio
Corn	control	$<1 \times 10^{-11}$	—	—
	sludge-amended	1.3×10^{-11}	39%	2.9×10^{-5}
Turnips	control	2.1×10^{-11}	58%	8.8×10^{-3}
	sludge-amended	3.8×10^{-11}	50%	8.9×10^{-5}
Broccoli	control	3.9×10^{-11}	36%	1.6×10^{-2}
	sludge-amended	6.8×10^{-11}	23%	1.6×10^{-4}
Tomato	control	2.5×10^{-11}	58%	1.0×10^{-2}
	sludge-amended	3.3×10^{-11}	43%	7.6×10^{-5}
Melon	control	$<1 \times 10^{-11}$	—	—
Cucumber	sludge-amended	$<1 \times 10^{-11}$	—	—

FIGURE CAPTIONS

Fig. 1. Location of high-volume cascade impactor downwind from experimental garden plot.

Fig. 2. Particle size collection characteristics of Anderson high-volume cascade impactor.

Fig. 3. Schematic of parallel-flow dissolution system and filter holder assembly.

Fig. 4. Organ burdens as a function of time following hypothetical exposure.

Fig. 5. Integrated organ doses as a function of time following hypothetical exposure.

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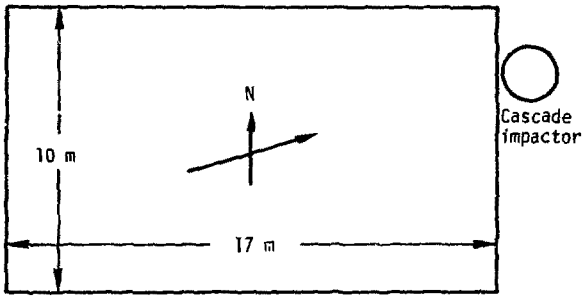

Fig. 1. Location of high-volume cascade impactor downwind from experimental garden plot.

Myers - Fig. 1

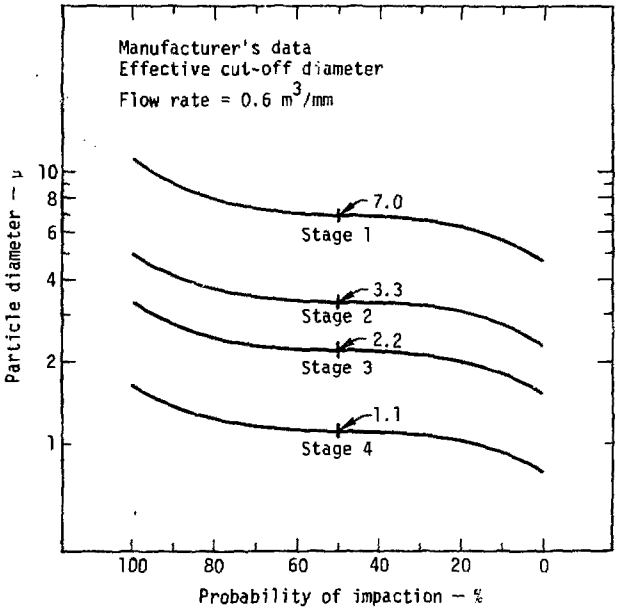

Fig. 2. Particle size collection characteristics of Anderson high-volume cascade impactor.

Myers - Fig. 2

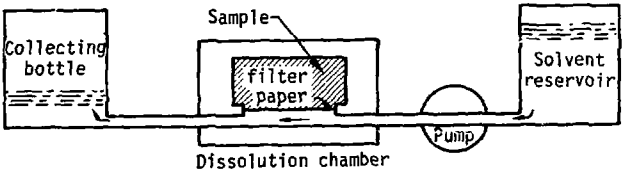

Fig. 3. Schematic of parallel-flow dissolution system and filter holder assembly.

Myers - Fig. 3

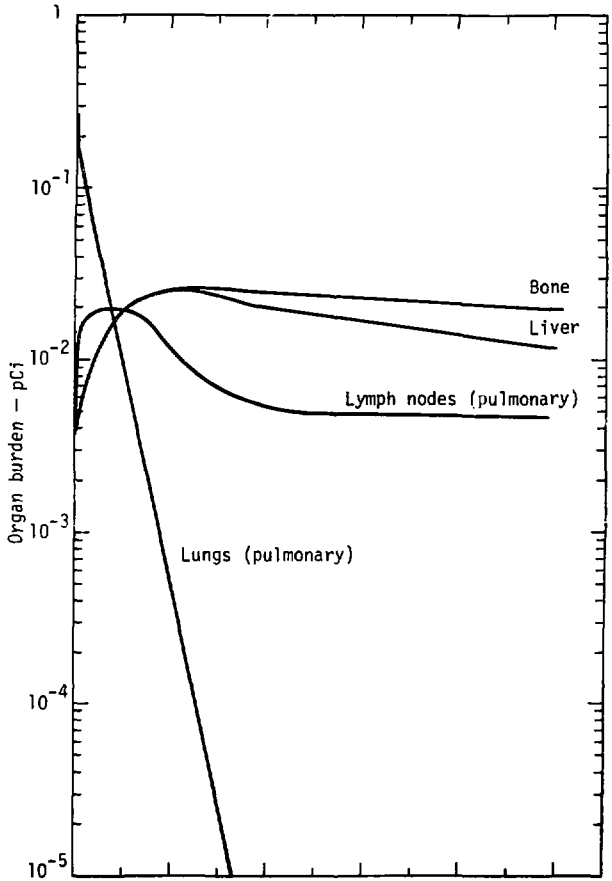

Fig. 4. Organ burdens as a function of time following hypothetical exposure.

Myers - Fig. 4

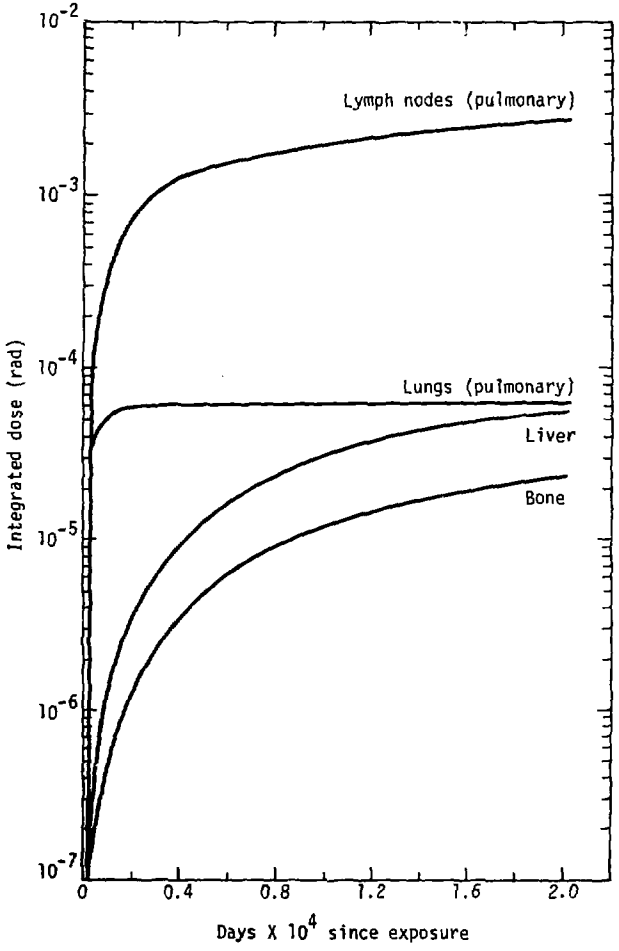

Fig. 5. Integrated organ doses as a function of time following hypothetical exposure.

Myers - Fig. 5